Dimensionless Groups For Understanding
Free Surface Flows of Complex Fluids

Gareth H. McKinley
Director, Hatsopoulos Microfluids Laboratory
Dept. Mechanical Engineering, M.I.T.
Cambridge MA 02139

Numerous processing operations of complex fluids involve free surface deformations; examples include spraying and atomization of fertilizers and pesticides, fiber-spinning operations, paint application, roll-coating of adhesives and food processing operations such as container- and bottle-filling. Systematically understanding such flows can be extremely difficult because of the large number of different forces that may be involved; including capillarity, viscosity, inertia, gravity as well as the additional stresses resulting from the extensional deformation of the microstructure within the fluid. Consequently many free-surface phenomena are described by heuristic and poorly-quantified words such as 'spinnability', 'tackiness' and 'stringiness'. Additional specialized terms used in other industries include 'pituity' in lubricious aqueous coatings, 'body' and 'length' in the printing ink business, 'ropiness' in yogurts and 'long/short textures' in starch processing.

A good approach to systematically getting to grips with such problems is through the tools of dimensional analysis (Bridgman, 1963) and this short note is intended to review the physics behind some of the plethora of specialized dimensionless groups one encounters when reading the scientific literature. The dominant balance of forces controlling the dynamics of any process depends on the relative magnitudes of each underlying physical effect entering the set of governing equations. In many (but certainly not all) problems, the dynamics are controlled by no more than two or three different contributions; and results from disparate experimental observations or numerical studies which describe different processing regimes or domains of stable/unstable operation can often be conveniently assembled in terms of processing diagrams such as those sketched in Figures 1 & 2.

For bulk flows of non-Newtonian fluids the relative importance of inertial effects and elastic stresses with respect to viscous stresses are characterized by the *Reynolds*

number, $Re = \rho V \ell / \eta_0$ and the *Weissenberg number* $Wi = \lambda V / \ell$ respectively. Here V and ℓ are characteristic velocity and length scales for the flow of interest and ρ, η_0, λ are the density, viscosity and longest – or characteristic – relaxation time of the test fluid. I have chosen the notation *Wi* here to avoid confusion with the *Weber number* which is commonly used in free surface flows (see further discussion below); however, much of the older literature will use *We* or *Ws* for this dimensionless ratio of elastic to viscous stresses in a deforming viscoelastic fluid.
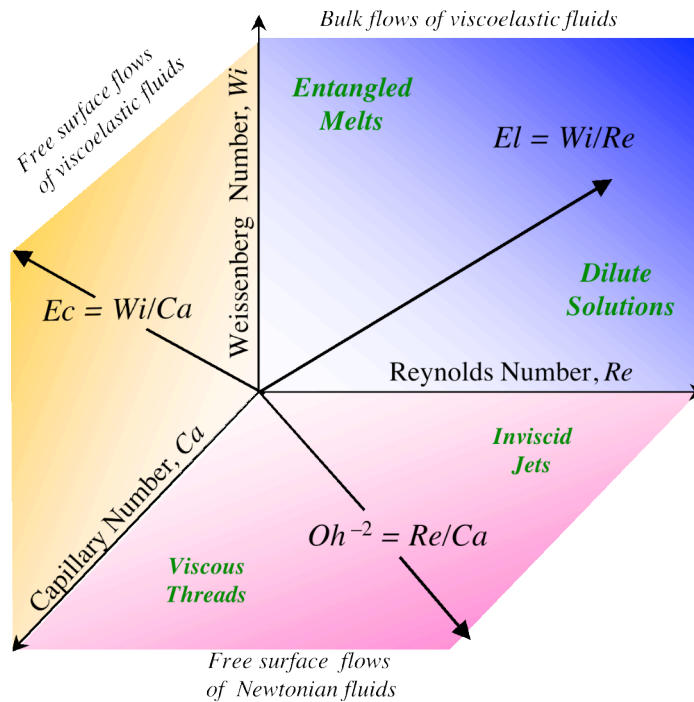
Figure 1: 'Operating diagram' showing the key dimensionless parameters characterizing free surface flows of complex fluids.

The product $\lambda V / \ell$ may also be interpreted as a Deborah number (Reiner, 1964) as it is a ratio of the polymeric time scale to a convective timescale $t_{conv} = \ell / V$. Indeed, in a steady channel flow the Deborah number and the Weissenberg number are interchangeable. However, as first noted by Metzner et al. (1965), there are important differences in more complex flows that are associated with the unsteadiness (in either the Eulerian or Lagrangian sense) of the process, and these two dimensionless measures of viscoelastic effects are typically interconnected by a dimensionless geometric parameter

characterizing the process of interest. The debate around the most appropriate terminology is endless and not terribly helpful (see Metzner et al. (1965) and Bird et al. (1987) Chp. 2 for further discussion); here we shall retain the notation *Wi* because we use the Deborah number below to describe a true ratio of timescales that are of importance in free surface flows

Prototypical steady polymer processing operations can be compared by their relative location in the back-plane of Figure 1. If we consider a particular process operation as a set of coordinates in this operating space we thus move outwards, away from the origin as the process throughput (i.e. the characteristic velocity *V*) is incremented and both the Reynolds number and the Weissenberg increase. The relative magnitudes of inertial stresses and elastic stresses can also be compared by taking the ratio of the two groups represented on the two axes. This parameter is not associated with any particular famous fluid mechanician or rheologist but is now commonly referred to as the *elasticity number* (Denn and Porteous, 1971) or sometimes the first elasticity number (Astarita and Marrucci, 1974). From the definitions of *Re* and *Wi* we obtain

$$\text{Elasticity Number:} \qquad El \equiv Wi \big/ Re = \eta_0 \, \lambda \big/ \rho \ell^2 \qquad (1)$$

It can be seen that this group is independent of the process kinematics (i.e. *V)* and only depends on the fluid properties and the geometry of interest. It thus sets the slope of the trajectory followed through the processing diagram and corresponds to the ratio of the time scale for stress evolution (λ) to the time scale for diffusion of momentum ($\ell^2 \big/ v$, where $v \equiv \eta_0 / \rho$ is the kinematic viscosity of the fluid). For example, extrusion of polymer melts corresponds to *El* >> 1, whereas processing flows for dilute polymer solutions (such as spin-casting) typically correspond to *El* << 1.

In analogous fashion, free surface flows of Newtonian fluids can be characterized by the magnitude of the Reynolds number and the capillary number $Ca = \eta_0 V / \sigma$ (where σ is the surface tension of the fluid), and can be represented on the horizontal plane of Figure 1. The slope of trajectories in this plane is again independent of the imposed velocity and corresponds to differing values of a parameter now referred to as the *Ohnesorge number* (Ohnesorge, 1936). There is much inconsistency in the use of this group; in his original work on jet breakup, von Ohnesorge defined this dimensionless parameter by the term $Z = \eta_0 \big/ \sqrt{\rho \sigma \ell}$. However, it is more common nowadays to use the

notation *Oh*, and the trajectory of interest is given by the <u>inverse square</u> of the Ohnesorge number, $Oh^{-2} \equiv Re/Ca = (\rho\sigma\ell)/{\eta_0}^2$. This group may also be usefully viewed as a Reynolds number based on a characteristic 'capillary velocity' $V_{cap} = \sigma/\eta_0$ (i.e. the velocity at which a viscous thread of fluid such as glycerol or pancake syrup would thin down and break; see for example Eggers (1997)). Coating operations and jetting/spraying operations with Newtonian fluids are fully parameterized by the values of *Ca* and *Oh* for the process of interest (for specific results see Quéré (1999) and Basaran (2002) respectively). However other combinations of these dimensionless parameters may also be used; for example, studies of high-speed jet breakup are commonly reported in terms of the *Weber number* $We = Re\,Ca = \rho V^2 \ell/\sigma$ and the Ohnesorge number (Becher, 1990).

Inertialess flows of elastic fluids with a free surface are represented by the left-hand vertical plane. Taking the ratio of the abscissa and ordinate axes gives another dimensionless parameter measuring the combined importance of elastic and capillary effects as compared to viscous stresses. This parameter can thus be thought of as an *elasto-capillary number* $Ec \equiv Wi/Ca = \lambda\sigma/(\eta_0\ell)$ and it is again a function only of the fluid rheology and the geometry. Much less is known about flows in this plane. Bousfield et al. (1986) were the first to study the nonlinear evolution of viscoelastic fluid jets and showed that increasing the elastocapillary number (denoted ϕ therein) resulted in strong stabilization of the jet. More recently, Spiegelberg and McKinley (1996) and Rasmussen and Hassager (2001) have investigated the effects of changes in *Ec* on adhesive fingering instabilities.

The three-dimensional interior of the parameter space shown in Figure 1 corresponds to general visco-elasto-capillary flows. The relative co-ordinates of a particular process or geometry can then be specified by values of $\{Re, Ca, Wi\}$. Since all three of these parameters vary with the characteristic process speed *V*, it is preferable to pick a single dynamical variable (say *Re*) that scales with *V* and then specify the other two coordinates using the material/geometric parameters, *Oh* and *El*. The literature in this area is much less well-developed and a fertile area for future research.

One particularly important subclass of free-surface flows are 'self-thinning' processes in which no external driving force is imposed (or in which the internal dynamics that develop spontaneously in the fluid are much faster than any external forcing). In such flows the fluid jet, thread, film or sheet thins down and breaks up

naturally under the action of capillary forces. To characterize this breakup process it is then necessary to seek a different set of dimensionless parameters that do not depend on the velocity scale V. Such capillary-thinning and break-up processes are governed by (at least) three characteristic time scales; a viscous time scale $t_{visc} \sim \eta_0 \ell / \sigma$, the polymeric timescale $t_{poly} \sim \lambda$ and an inertial or Rayleigh time-scale $t_R \sim \sqrt{\rho \ell^3 / \sigma}$ (Rayleigh, 1879). The relative balances of these time-scales (and the associated contributions to the total force in the fluid thread) can thus be represented in terms of two dimensionless parameters; one of these is the Ohnesorge Number we have seen before; $Oh = t_{visc}/t_R \sim \eta_0 / \sqrt{\rho \sigma \ell}$. The other parameter is the ratio of the polymeric relaxation time to the Rayleigh time-scale for capillary breakup. This parameter corresponds to an 'intrinsic' or 'natural' Deborah number for free surface viscoelastic flows and represents the ratio of the time scale for elastic stress relaxation, to the 'Rayleigh time scale' for inertio-capillary break-up of an inviscid jet:

$$\text{Intrinsic Deborah Number;} \qquad De = \frac{t_{polym}}{t_R} = \frac{\lambda}{\sqrt{\rho \ell^3 / \sigma}} \qquad\qquad (2)$$

Note that in contrast to the Weissenberg number defined above, this viscoelastic parameter does not depend on the imposed kinematics (*V*). These two dimensionless parameters can be used to define a two-dimensional 'operating space' for instruments such as capillary-thinning/break-up extensional rheometers and other self-thinning free surface processes involving non-Newtonian fluids. Such a diagram is sketched in Figure 2 below.
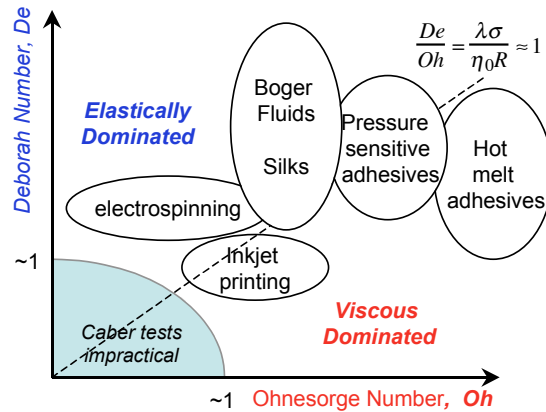

Figure 2   An 'operating diagram' for capillary self-thinning and break-up of

Successful operation of a capillary thinning instrument or other non-Newtonian free surface flow typically requires that the fluid is 'sufficiently viscous' or has enough 'spinnability'. Diagrams such as Figure 2 allow us to express such verbal statements in terms of dimensionless parameters. For example in a capillary thinning test we require surface tension to drive the thinning and either viscosity or elasticity to resist the necking process; therefore we need either $Oh \gtrsim 1$ or $De \gtrsim 1$ (Rodd et al., 2005). A 'low viscosity elastic' fluid (Harrison and Boger, 2000) corresponds (at least in a free surface flow) to $t_{polym} > t_R > t_{visc}$ or equivalently $De > 1 > Oh$. In such flows, viscous effects are never important (see, for example, Amarouchene *et al.* (2001)) and one is interested in the limit of 'inviscid elastic fluids' or more accurately irrotational flows of elastic liquids (Wang et al., 2005). Similarly, in operations such as electrospinning it is essential to be able to form a uniform filament in the absence of beads-on-a-string morphology (Fong et al., 1999); hence one needs to design the fluid rheology and geometry to ensure $De \geq Oh > 1$. Conversely in inkjet printing neither long elastic tails nor inertially-induced satellite drops are desirable; consequently one may seek $De \approx Oh \approx 1$.

For very viscous fluids, the inertial time-scale t_R of course becomes irrelevant and the dynamics of the thinning process depends solely on the relative magnitude of the viscous and elastic time scales. This leads us again to the elasto-capillary number

$$\text{Elastocapillary Number} \qquad Ec = \frac{De}{Oh} = \frac{t_{poly}}{t_v} = \frac{\lambda\sigma}{\eta_0 \ell} . \qquad (3)$$

Elastically-dominated and viscously-dominated self-thinning and necking processes (for example the fibrils that form during decohesion of tacky polymer melts and solutions (Rasmussen and Hassager, 2001) are thus demarcated by the line $E \approx 1$ sketched in Figure 2.

In this short note I have attempted to provide an overview of some of the spectrum of dimensionless groups used in describing free surface flows of elastic fluids. I have not discussed the perturbative effects of gravity explicitly. Consideration of the gravitational body force leads to a characteristic length scale referred to as the *capillary*

length $\ell_{cap} \sim \sqrt{\sigma/\rho g}$. For most fluids $\ell_{cap} \approx$ 1-2mm, and on length scales smaller than this gravity becomes increasingly unimportant. This length scale can also be substituted into any of the dimensionless groups defined above to lead to yet another set of dimensionless groups! It is worth noting that as interest in microscale and nanoscale manufacturing intensifies, and the characteristic length-scale of a particular process decreases, the elasticity number, elastocapillary number and intrinsic Deborah number will all increase. Non-Newtonian effects in the processing of complex fluids will thus become increasingly prevalent on the microscale. For example, in 'low viscosity' and 'weakly elastic' fluids, inertial effects often overwhelm the non-Newtonian stresses in the fluid making both the shear and extensional rheology difficult to measure (see e.g. Linder *et al*. (2003)). A consideration of the dimensionless parameters defined above shows that as the length scale ℓ decreases into the micrometer range, inertial forces in a fluid thread become progressively less important and viscoelastic forces once again dominate. Microscale measurements using instruments such as capillary-thinning devices may thus make excellent micro-rheometers for 'low viscosity non-Newtonian fluids' such as agricultural fertilizers, and inks or paints used in jetting and spraying operations.

Bibliography

Amarouchene, Y., Bonn, D., Meunier, J. and Kellay, H., Inhibition of the Finite Time Singularity during Droplet Fission of a Polymeric Fluid, *Phys. Rev. Lett*., **86**(16), (2001), 3558-2562.

Astarita, G. and Marrucci, G., <u>Principles of Non-Newtonian Fluid Mechanics</u>, McGraw-Hill, Maidenhead (UK), 1974.

Basaran, O., Small-Scale Free Surface Flows with Breakup: Drop Formation and Emerging Applications, *A.I.Ch.E. J*., **48**(9), (2002), 1842-1848.

Becher, P., The so-called Ohnesorge Equation, *J. Coll. Int. Sci*., **140**(1), (1990), 300-301.

Bird, R.B., Armstrong, R.C. and Hassager, O., <u>Dynamics of Polymeric Liquids. Volume 1: Fluid Mechanics</u>, Vol. 1, 2nd Edition, Wiley Interscience, New York, 1987.

Bousfield, D.W., Keunings, R., Marrucci, G. and Denn, M.M., Nonlinear Analysis of the Surface-Tension Driven Breakup of Viscoelastic Fluid Filaments, *J. Non-Newt. Fluid Mech*., **21**, (1986), 79-97.

Bridgman, P.W., <u>Dimensional Analysis</u>, Yale Paperbound, New Haven, 1963.

Denn, M.M. and Porteous, K.C., Elastic Effects in Flow of Viscoelastic Liquids, *Chem. Eng. J.*, **2**, (1971), 280-286.

Eggers, J., Nonlinear Dynamics and Breakup of Free-Surface Flows, *Rev. Mod. Phys.*, **69**(3), (1997), 865-929.

Fong, H., Chun, I. and Reneker, D.H., Beaded Nanofibers Formed During Electrospinning, *Polymer*, **40**, (1999), 4585-4592.

Harrison, G.M. and Boger, D.V., Well-Characterized Low Viscosity Elastic Liquids, *Appl. Rheol.*, **10**(4: July/Aug), (2000), 166-177.

Lindner, A., Vermant, J. and Bonn, D., How to obtain the Elongational Viscosity of Dilute Polymer Solutions, *Physica A*, **319**, (2003), 125-133.

Metzner, A., White, J.L. and Denn, M.M., Constitutive Equations for Viscoelastic Fluids for Short Deformation Periods and for Rapidly Changing Flows: Significance of the Deborah Number, *A.I.Ch.E. J.*, **12**, (1965), 863-866.

Ohnesorge, W.v., Die Bildung von Tropfen an Düsen und die Auflösung flüssiger Strahlen, *Z.A.M.M.*, **16**(6), (1936), 355-358.

Quéré, D., Coating of a Fiber, *Ann. Rev. Fluid. Mech.*, **31**, (1999), 347-384.

Rasmussen, H.K. and Hassager, O., The Role of Surface Tension on the Elastic Decohesion of Polymeric Filaments, *J. Rheol.*, **45**(2), (2001), 527-537.

Rayleigh, L., On the Instability of Jets, *Proc. Lond. Math. Soc.*, **10**, (1879), 4-13.

Reiner, M., The Deborah Number, *Physics Today*, (1964), 46.

Rodd, L.E., Scott, T.P., Cooper-White, J.J. and McKinley, G.H., Capillary Breakup Rheometry of Low-Viscosity Elastic Fluids, *Appl. Rheol.*, **15**(1), (2005), 12-27.

Spiegelberg, S.H. and McKinley, G.H., Stress Relaxation and Elastic Decohesion of Viscoelastic Polymer Solutions in Extensional Flow, *J. Non-Newtonian Fluid Mech.*, **67**, (1996), 49-76.

Wang, J., Joseph, D.D. and Funada, T., Purely Irrotational theories of the Effects of Viscosity and Viscoelasticity on Capillary Instability of a Liquid Cylinder, *J. Non-Newt. Fluid Mech.*, (2005), in press.